Thick-Film Thermoelectric Microdevices

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Abstract

Miniaturized thermoelectric devices integrated into thermal management packages and low power, high voltage, electrical power source systems are of interest for a variety of space and terrestrial applications. In spite of their relatively low energy conversion efficiency, solid-state microcoolers microgenerators based on state-of-the-art materials offer attractive solutions to the accelerating trend towards miniaturization of electronic components and "system on a chip" concepts where the functions of sense, compute, actuate, control, communicate and power are integrated. The miniaturization of state-of-the-art thermoelectric module technology based on Bi₂Te₃ alloys is severely limited due to mechanical and manufacturing constraints. Compared to bulk technology, the key advantages of integrated microdevices designed with thousands of thermocouples are their ability to handle much higher heat fluxes (thus resulting in high power densities), their much faster response time as well as the possibility of generating high voltages under small temperature differentials. We are currently developing novel microdevices with a conventional vertically integrated configuration combining high thermal conductivity substrates such as diamond or silicon, integrated circuit technology, and electrochemical deposition of thick thermoelectric films. We report here on our progress in developing techniques for obtaining 10-50 µm thick films of p- and n-type Bi₂Te₃ alloys by electroplating through a thick photoresist template on top of patterned multilayer metallizations. This microdevice fabrication technology is now being developed for several applications, including a high cooling power density microcooler (200 W/cm²) for thermal management of power electronics and a 100mW autonomous hybrid thermoelectricrechargeable batteries generator using low grade waste heat. Future directions of research are also discussed.

Introduction

Solid state thermoelectric devices have demonstrated attractive characteristics such as long life, the absence of moving parts or emissions, low maintenance and high reliability. In spite of a large number of potential civilian and military applications, their use has been severely limited due to their relatively low energy conversion efficiency and high development costs. To broaden the field of thermoelectrics, higher performance devices and systems need to be developed. One approach to achieve this goal is the discovery and infusion of novel thermoelectric materials more efficient than the current state-of-the-art Bi-Sb, Bi₂Te₃, PbTe or Si-Ge alloys. Recent results in several laboratories have successfully identified superior materials in several temperature ranges [1-3]. There is currently an effort to introduce some of these new compounds into simple

unicouple power generator configurations to demonstrate the increased conversion efficiency [4].

A second approach is to significantly improve the design, specific power (watts per unit area or volume) and lower the costs of thermoelectric devices even when using state-of-the-art thermoelectric materials. A key feature of thermoelectrics is its scalability, as illustrated in equation (1) and Figure 1 for a refrigerator device. Consequently, miniaturized devices based on Bi₂Te₃ alloys and integrated into thermal management packages and low power, high voltage, electrical power source systems are quite attractive for a variety of space and terrestrial applications.

$$Q_{cold}^{\max} = \frac{A}{l} \left[\frac{1}{2} \frac{S_{pn}^2 T_{cold}^2}{\rho_{pn}} - \lambda_{pn} \left(T_{hot} - T_{cold} \right) \right]$$
(1)

Where Q_{cold}^{\max} , T_{hob} T_{cold} S_{pm} ρ_{pm} λ_{pm} A, and l are respectively the maximum cooling power, hot junction and cold junction temperatures, Seebeck coefficient, electrical resistivity, thermal conductivity, cross-sectional area and length of a p-n thermoelectric leg couple. Note that Q_{cold}^{\max} is directly proportional to the A/l leg geometric aspect ratio.

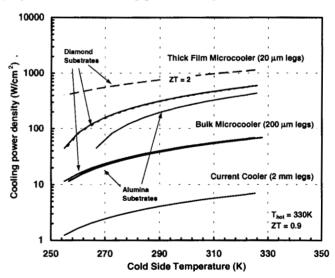
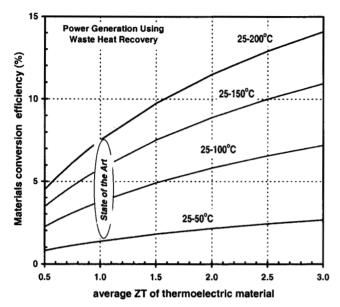
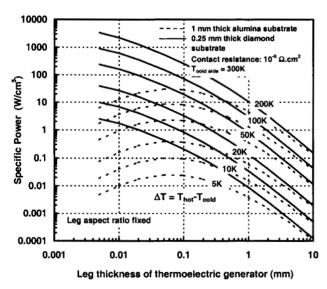


Figure 1: Cooling power densities as a function of the temperature differential across increasingly miniaturized thermoelectric cooler configurations (A/I constant). For thick film devices operating under high heat flux densities, high thermal conductivity substrates will minimize performance degradation due to heat losses. The impact of improving the materials figure of merit ZT from 0.9 to 2.0 is also shown.

In spite of their relatively low energy conversion efficiency, solid-state microcoolers and microgenerators based on state-of-the-art materials offer attractive solutions to the accelerating trend towards miniaturization of electronic components and "system on a chip" concepts where the functions of sense, compute, actuate, control, communicate and power are located together. The drive for increased performance and miniaturization of a wide range of electronic systems requires higher power levels, higher packaging densities and faster response time.



<u>Figure 2</u>: Calculated improvement in thermoelectric generator performance when using more efficient materials. The temperature differences reported here are relevant to state-of-the-art Bi₂Te₃ alloys and miniaturized devices. Substantially higher conversion efficiencies can be achieved by operating across a wider temperature range.



<u>Figure 3</u>: Calculated increase in specific power (per unit volume) with increasing leg miniaturization (constant A/I ratio) and increasing temperature differential of operation. Data are shown for both low (alumina, blue dots) and high (diamond, red lines) thermal conductivity substrates. The calculation for increasingly miniaturized devices implies a corresponding increase in heat flux density delivered to the hot junction side of the device.

Thermal management problems have now become a major issue for this technological process because they limit the degree of integration of devices and components [5]. In addition to reliability issues, significant performance improvement can be obtained by operating the active junction of semiconductor chips at temperatures near or lower than ambient, as well as by maintaining precise temperature control. This is especially true of several electronic and photonic devices such as microprocessors, power amplifiers and infrared lasers [6, 7]. For both aerospace and terrestrial applications, there is also a growing need for developing miniaturized on-chip low power improved batteries with high specific power, long life, high voltage, resistance to extreme temperatures, and low environmental impact characteristics [8, 9]. Figures 2 and 3 illustrate the potential performance of miniaturized thermoelectric generators in terms of energy conversion efficiency (Figure 2) and volume specific power (Figure 3).

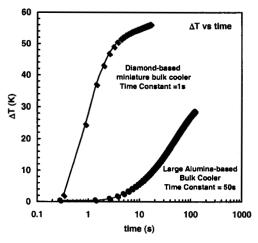
Device Miniaturization

Current thermoelectric module technology is ill suited to the development of miniaturized devices due to mechanical and manufacturing constraints for thermoelement dimensions (100-200µm thick minimum) and number (100-200 legs maximum). In addition to the widespread use of semi-manual assembly techniques that results in high costs for more compact configurations, these devices have typically undesirable high current and low voltage characteristics. Moreover, electrical contact resistances and heat transfer issues at the device level are typically not critical for large bulk thermoelectric modules but need to be addressed when considering device miniaturization [10].

For power generation, much smaller devices capable of high voltage (up to 5V) power output in the nW to tens of uW range have already been developed: monolithic structures and Most of the monolithic more recently thin film devices. module configurations have been used in nuclear battery type devices, operating across large temperature differences (100-200K), with a small amount of radioisotope material (usually PuO₂) as the heat source [8, 11]. The specific power density of the monolithic thermopiles is typically measured in tens of mW/cm³, but falls to about 60 μW/cm³ when taking into account the complete power source package. Thin film devices producing 20 mW at 4V under load with a temperature difference of 20K have been recently described [12]. The 0.22 cm³ device is comprised of 2250 thermocouples deposited on Kapton thin foils packed together and was fabricated using integrated circuit-type techniques. However, in spite of this remarkable achievement that could allow for batch fabrication of these devices, the specific power density still remains quite low, close to 90 µW/cm³ (heat source not included). This is mainly due to the fact that the length of the thermoelectric legs is supported by the Kapton substrate, thus introducing a very significant thermal shunt and dramatically degrading conversion efficiency.

There has been some effort at miniaturizing thermoelectric cooling devices with activities concentrating on fast response time and ultra-stable temperature control. Experimental results and detailed analyses of the performance of miniature bulk coolers based on diamond substrates with 120 legs, 0.2

mm thick and 0.4x0.4 mm in cross-section have been reported [13,14]. In addition to being able to reach maximum temperature differentials comparable to those achieved in much larger bulk coolers, such miniature devices have demonstrated their ability to handle larger heat flux densities and much shorter response time, as illustrated in Figure 4.



<u>Figure 4</u>: Improvement in response time to maximum DT for miniaturized bulk cooler based on 0.5mm thick diamond substrates and 0.2mm thick, 0.4x0.4mm cross-section legs. Experimental data was obtained at 293K.

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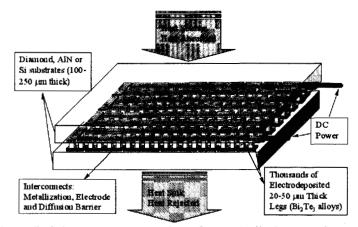
To circumvent key shortcomings of the current technology described in the preceding section, the Jet Propulsion Laboratory (JPL) is pursuing the development of vertically integrated thermoelectric microdevices that can be fabricated using a combination of thick film electrochemical (ECD) and integrated circuit (IC) processing techniques [15].

Microdevice configuration

The term "vertically integrated" here refers to the conventional thermoelectric module configuration shown in Figure 5. This design eliminates the large heat losses observed in planar thin film thermoelectric devices where the legs are deposited onto a supporting substrate and the heat flow is parallel to the substrate. However, planar configurations do offer a very convenient way of fabricating electrical interconnects between the thin film legs by using traditional masking techniques. Thermal resistances due to heat transfer through the metallizations and substrates, as well as electrical resistances due to the interconnects between ntype and p-type thermoelectric legs, rapidly become important issues when increasing device miniaturization. High thermal conductivity substrates, thin metallizations and intimate contact with the heat source and heat sink media are key to minimizing thermal issues, in particular when the microdevices operate under high heat flux conditions.

For example, in the case of microgenerators, since high voltage power output are highly desirable from a power conditioning aspect, this means that the microdevices will typically possess several thousands of very short thermocouples. Electrical contact resistances can thus easily become a very large fraction of the total internal device resistance. However, low values are routinely obtained in the

electronic semiconductor industry and similar processing techniques have been developed here. Finally thermally stable diffusion barriers are needed to maintain the integrity of the multilayered stack of substrates, metallic interconnects and thermocouples. The effectiveness of amorphous transition metal nitride diffusion barriers for metallizations on diamond, AlN and thermally oxidized silicon substrates has been recently demonstrated [16].



<u>Figure 5</u>: Schematic representation of a vertically integrated thick film thermoelectric microgenerator using thin high thermal conductivity substrates.

Microdevice fabrication

Electrodeposition

Hot side temperatures for microdevice applications that we are currently considering are 200 to 500K. Bi_{2-x}Sb_xTe₃. _vSe_v alloys are the state-of-the-art materials best suited to these temperatures of operation. Since the thickness of the legs selected in our various device concepts ranges from 10 to 60µm, we have actively pursued the development of an electrochemical thick film deposition process. ECD constitutes an inexpensive way to synthesize semiconducting films [17] and, depending on the current density used in deposition, the deposition rate can be varied widely, up to several tens of microns per hour. In addition, slight variations in the deposition potential or solution concentration may possibly be used to induce off-stoichiometric films, thus providing p- or n-type doping through stoichiometric deviation. The electrodeposition of thermoelectric materials has not been widely investigated [18, 19] and new experimental methods have been developed to obtain p-type and n-type Bi_{2-x}Sb_xTe_{3-y}Se_y compositions which are optimal for thermoelectric power generation in the temperature range of interest. An additional advantage of ECD is that some of the interconnect layers necessary to the fabrication of these devices, such as Cu for the electrical path or Ni for the Cu diffusion barrier can also be deposited by using different aqueous solutions.

Depositions are typically run near room temperature using standard electrochemistry techniques: a three electrode cell with open beaker configuration but with separate vessels for the reference electrode (saturated calomel electrode, SCE) and the counter/working electrodes. A salt bridge is used to electrically connect the two beakers. The counter electrode consists of a fine Pt mesh while metallic foils or metallized high thermal conductivity substrates such as diamond, AlN or Si/SiO₂ are

used for a working electrode. Solutions contain dissolved high purity elements (Bi, Sb, Te, Se) into an acidic aqueous medium, typically HNO₃ and deionized water (pH ~ 0). Concentration of the elements in the electrolyte is typically varied between 0.0001 and 0.01 M. In the case of Sb-rich p-type Bi_{2-x}Sb_xTe₃ films where 1.3<x<1.5, a chelating agent must be added to prevent the spontaneous precipitation of an insoluble oxide compound and raise its maximum solubility (about 8x10⁻⁴ M in 1 M HNO₃ aqueous solution). Both the electrodeposition and cyclic voltammetry measurements are carried out using mechanical solution stirring and a computer-controlled EG&G Princeton Potentiostat/ Galvanostat 273A. Experimental results have demonstrated that both n-type and p-type Bi₂Te₃ alloy films can be deposited with transport properties similar to those of bulk Deposition rates typically range from 1 to materials. 15µm/hour, depending on electrolyte concentrations in Bi, Sb, Te or Se and applied voltage. Deposition rates are typically slower for p-type films due to the chelating additives. More details have been reported elsewhere [15].



20µm

<u>Figure 6</u>: Thick positive photoresist template on top of a metallized Si/SiO₂ substrate. Deep cylindrical holes where the thermoelectric leg will be deposited can be seen.

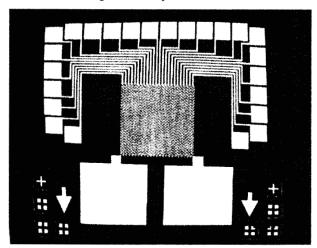
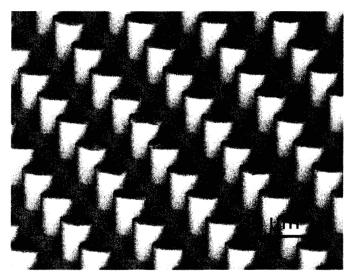


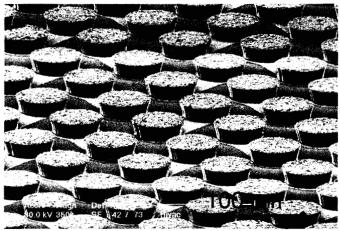
Figure 7: Cu metallization on top of a Si/SiO₂ substrate where interconnects have been patterned for subsequent deposition of the thick photoresist template and thermoelectric legs. The fully metallized square pads are for providing electrical contact tests.

Integrated Circuit Processing Approach

Building on the availability of new thick photoresist commercial products, we have developed templates suitable to the electrochemical deposition of legs as thick as $70\mu m$ and as small as $6\mu m$ in diameter. Actually, it has been determined that to be able to tightly control the geometry of the legs and prevent "mushrooming" growth, electrodeposition must be conducted in equally thick photoresist templates. The thick positive photoresist template is patterned with deep square or round shaped holes that must be pre-aligned on top of metallic interconnects. Figures 6 and 7 illustrate the result of IC-type processing.



<u>Figure 8</u>: Bi₂Te₃ thermoelectric legs deposited on a metallized Si substrate using a thick photoresist template and throughmask plating technique.



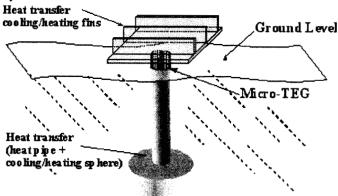
<u>Figure 9</u>: Bi₂Te₃ legs electrodeposited on top of Cu/Ni interconnect metallizations (using a Si/SiO₂ substrate).

More processing steps are required to successively deposit n-type and p-type legs on top of the bottom substrate interconnects (Figure 8 and 9), and then ensure proper joining to a top substrate with similarly patterned electrical interconnects. Based on commercial electrolytes, we have used ECD techniques to deposit high quality Cu, Ni and Pb-Sn solder layers as well. The Pb-Sn layer can be used to form solder bumps on top of the legs, as done for flip-chip bonding

techniques [20]. These processing steps are illustrated in Figure 9. The combination of ECD and IC-type techniques offers a degree of flexibility in designing and fabricating thermoelectric microdevices. A single photolithography mask can usually combine all of the necessary patterns to completely fabricate one microdevice configuration.

Microgenerators for "Energy Harvesting" Applications

To better illustrate possible applications of miniaturized thermoelectric generators, let us consider an "energy harvesting" scheme where an environmental heat source is used to provide electrical power to operate electronic components in remote or unattended locations. One potential compact power source configuration is represented in Figure 10 where a microgenerator makes use of the temperature difference between air and soil temperatures. microgenerator is used to trickle charge a set of rechargeable batteries or capacitors so that the hybrid power source can deliver a much higher power level for brief periods of time. Electronic measurement or communication devices coupled to the power source are thus allowed to operate following some nominal duty cycle. In this energy-harvesting scheme, thanks the expected reliability of the thermoelectric microgenerator, the electronics will be operational for as long as sufficient air/soil temperature differentials exist and the energy storage components can handle the charge/discharge cycles.



<u>Figure 10</u>: Schematic representation of an environmental energy harvesting power source concept that could be used to operate low power electronic components and eliminate the need for conventional batteries.

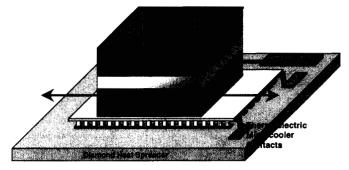
This hybrid system consists of a small $6x6 \text{ cm}^2$ aluminum box covered with two layers of fins that is located at the surface while the heat is transferred to the soil by means of a 30cm long heat pipe terminated by a small spherical or cylindrical heat exchanger. Preliminary design operating conditions only require a low wind speed of 0.75 m/s and air and soil temperatures of respectively 300K and 287K. Calculations show that the effective temperature difference across the thermoelectric microgenerator would be only 8.5 K, but would be sufficient to produce about 22 mW of power at 4.1 V under resistive load. The microgenerator configuration is a set of 2300 n-type and p-type leg couples $50 \mu \text{m}$ in thickness. The specific power performance of the device under such operating conditions is about 1.3W/cm^3 with a conversion efficiency of 0.4%.

However, temperatures will fluctuate during the day and at night the temperature profile will be reversed as the air becomes colder than the soil. As long as a temperature differential will be maintained across the thermoelectric microdevice, power will be generated. Power conditioning electronics are needed to handle the variations in output power, voltage and current over time and ensure proper charging and discharging of the energy storage components. Based on the design operating conditions described here, we estimate that the hybrid power source could deliver 100mW following a 10% duty cycle.

The first experimental characterization of the performance of various vertically integrated microgenerator devices will be carried out in the very near future at JPL as final assembly steps are now being developed. Results will be reported in later publications. If successful, we expect that this approach could be applied to a number of heat sources with different temperature ranges (such as combustion processes) and for a variety of power levels.

Microcoolers for Mid-IR Lasers

Another application area of interest for microdevices is integrated thermal management of optoelectronics such as mid-IR lasers. Mid-infrared lasers have many potential military and commercial applications such as detection of trace gases and emission controls. Unfortunately, these lasers are limited to cryogenic temperatures for CW and quasi-CW operation, requiring liquid cryogen or bulky and expensive cryogenic coolers. New quantum and interband cascade lasers have reached pulsed-mode room-temperature operation, but very low duty cycles needed to limit device heating strongly limit their performance [21].



<u>Figure 11</u>: Schematic representation of a thermoelectric microcooler monolithically integrated with the active region of a mid-IR laser (cold junction) and a diamond heat spreader (hot junction).

The thermal management solution pursued by JPL in collaboration with several universities is a monolithically integrated thermoelectric microcooler for local transient cooling of the active area of mid-IR lasers, as shown in Figure 11. One of the key aspects of this effort is to study the use of transient cooling effects in such solid-state devices. More theoretical specifically, the maximum steady-state temperature drop that can be reached with the state-of-art thermoelectric materials is limited to about 80 K and for practical bulk devices maximum ΔT values are close to 70 K An additional temperature drop, however, can be realized by superimposing a transient current on the optimum

steady-state operation current. This additional transient cooling effect results from delaying the Joule heating from propagating to the cold junction. Numerous studies have been reported in the past on the transient cooling effect in bulk thermoelectric coolers [23-26]. For a square pulse, it has been shown that the maximum additional temperature drop is comparable to the steady-state temperature drop. It has long been realized, however, that this may not be the maximum limit of the transient heating effect. By using time dependent transient pulses, significantly higher temperature drop may be In fact, the minimum reported cold junction temperature of bulk TE coolers under pulse conditions is below 100 K [23]. Those experimental results where subjected to questions because there were no good temperature sensing techniques to further validate the experimental data. Figure 12 shows an example of experimental transient cooling effect for bulk TE coolers. One study used the transient effect for the cooling of light emitting diode and the power output from the LED doubled under pulsed operation compared to the case without the Peltier cooler [27].

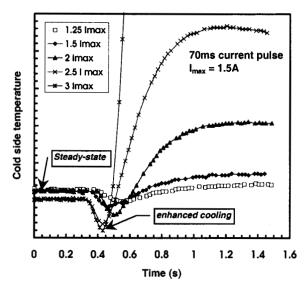


Figure 12: Transient cooling achievable from a commercially available thermoelectric cooler. Steady state operation at the optimal current provides a cold side temperature of about 230K. Increasing the current for 0.07s (pulse) first cools the cold side beyond that achievable with steady state operation. After some time, the additional joule heating from the pulse diffuses to the cold side, warming it above the steady state temperature. Using an optimized device geometry and pulse shape much larger transient cooling has been reported.

By combining the fast response time and localized enhanced transient cooling, optimally designed thermoelectric microcoolers could allow operating the mid-IR laser for much longer duty cycles at power levels practical for many applications.

Conclusion

Thermoelectric microdevices offer attractive solutions with high specific power output characteristics when considering low power electrical sources for remote or unattended electronics, and innovative integrated thermal

management solutions for electronic and optoelectronic components. However, thermoelectric technology to date is limited to bulky configurations based on monolithic thermopiles or to inefficient planar thin film devices. To fabricate high performance microdevices in a "classic" vertically integrated module configuration, a combination of electrochemical deposition techniques and integrated circuit technology is now under development. We have successfully electrodeposited both n-type and p-type thick Bi₂Te₃ alloy films (10-60 µm thick) from aqueous solutions and we have shown that transport properties similar to that of bulk materials can be achieved. Thermally stable metallizations to high thermal conductivity substrates and effective diffusion barriers for fabricating the electrical interconnects between the n- and p-type legs have also been demonstrated. Thick photoresist templates up to 70 µm have been successfully patterned developed and using conventional photolithography, resulting in the reproducible fabrication of highly packed arrays of thousands of legs as small as 6µm in We are now focusing on the fabrication of diameter. operational prototype devices to be used in several cooling and power generation applications.

Acknowledgments

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